

## **Translator's Declaration**

I, Seiki TAKITA residing at 30-5, Syakujii-dai 3-chome Nerima-ku, Tokyo, 177-0045 Japan, do hereby declare that I am conversant with the Japanese and English language and I further declare that the following translation made by me is to the best of my knowledge and believe it is a true and correct translation of Japanese Patent Application No. 1999/168864.

10th August, 2006



[Name of Document polication for patent

[Reference Number] 995003

[Filing Date]

June 15, 1999

[Addressee]

Commissioner of Japanese Patent Office

[International Patent Classification] C08K 5/527

[Inventor]

[Address]

c/o ASAHI DENKA KOGYO K. K.

2-13, Shirahata 5-chome, Urawa-shi,

Saitama Prefecture

[Name]

HARUNA Tohru

[Inventor]

[Address]

c/o ASAHI DENKA KOGYO K. K.

2-13, Shirahata 5-chome, Urawa-shi,

Saitama Prefecture

[Name]

TAKAHASHI Masayuki

[Applicant]

[Identification Number] 000000387

[Name]

ASAHI DENKA KOGYO K. K.

[Agent]

[Identification Number]

100090491

[Patent Attorney]

[Name]

MIURA Yoshikazu

[Indication of Fees]

[Prepayment Book Number]

026033

[Amount of Payment]

21000

[List of Submitted Articles]

[Name of Article] Specification

1

[Name of Article] Abstract 1
[General Power of Attorney Number] 9200692
[Necessity of Proof] Necessary

[Name of Document] Specification
[Title of Invention] Nucleating Agent
[Claims]

[Claim 1] A nucleating agent comprising a phosphoric acid aromatic ester metal salt having an average particle size of 10  $\mu$ m or less, an average aspect ratio of 10 or less, and a bulk specific gravity of at least 0.1, the metal salt being represented by the following formula (I):

[Chemical Formula 1]

$$\begin{bmatrix} R_1 & & & \\ R_2 & & & \\ R_3 & & P-O \\ R_2 & & & \\ R_1 & & & \end{bmatrix}_n A(OH)x \qquad (I)$$

(wherein  $R_1$  represents a C4-C8 alkyl group,  $R_2$  represents a hydrogen atom or a C1-C8 alkyl group,  $R_3$  represents a C1-C4 alkylidene group, A represents a metal having a valence of (n+x), n is an integer of 1 to 3, and x is an integer of 0 to 2).

[Detailed Description]

[0001]

[Industrial Field]

The present invention relates to a nucleating agent

containing a phosphoric acid ester metal salt, which exhibits excellent handling property and exerts excellent effects of improving transparency and mechanical strength of a resin; and more particularly to a nucleating agent characterized by having a certain average particle size, a certain aspect ratio, and a certain bulk specific gravity which individually fall within specified ranges, the agent exhibiting excellent handling property when incorporated into a resin, and exerting excellent effects of improving transparency and mechanical strength of the resin containing the agent.

[0002]

[Prior art]

Phosphoric acid aromatic ester metal salts are typical nucleating agents used to improve the crystallization temperature of a crystalline synthetic resin, transparency and mechanical strength of the resin. Such a phosphoric acid aromatic ester metal salt serving as a nucleating agent is a pillar-shaped crystalline compound. When such an agent is incorporated into a resin, in order to enhance dispersibility of the agent in the resin, the agent is preferably finely pulverized so as to attain a particle size of 10 µm or less. However, since a conventional finely pulverized nucleating agent has a large aspect ratio and a small bulk specific gravity, the nucleating agent is not suitable for automatic weighing. In addition, since the nucleating agent exhibits poor fluidity, the agent has the problem that transfer of the agent through a hopper, etc. is sometimes unsatisfactory.

[0003]

When a nucleating agent is finely pulverized, the dispersibility of the agent in a resin is enhanced, but the fluidity of the agent is lowered. In contrast, when the particle size of a nucleating agent is increased, the fluidity of the agent is enhanced, but the dispersibility of the agent in a resin is lowered, resulting in poor effects of improving transparency and mechanical strength of the resin. Therefore, there has been demand for a nucleating agent exhibiting excellent fluidity while assuming a fine particulate form.

[0004]

[Problems to be solved by the Invention]

In view of the foregoing, the present inventors have performed extensive studies, and have found that, when the average particle size, average aspect ratio, and bulk specific gravity of a phosphoric acid aromatic ester metal salt are controlled so as to fall within a specific range, the metal salt exhibits excellent fluidity while assuming a fine particulate form. The present invention has been accomplished on the basis of this finding.

[0005]

[Means to solve the Problem]

Accordingly, the present invention provides a nucleating agent comprising a phosphoric acid aromatic ester metal salt having an average particle size of 10  $\mu m$  or less, an average aspect ratio of 10 or less, and a bulk specific

gravity of at least 0.1, the metal salt being represented by the following formula (I):

[0006]

[Chemical Formula 2]

$$\begin{bmatrix} R_1 & & & \\ R_2 & & & \\ R_3 & & P-O \\ R_2 & & & \\ R_1 & & & \\ \end{bmatrix}_n A(OH)x \qquad (I)$$

(wherein  $R_1$  represents a C4-C8 alkyl group,  $R_2$  represents a hydrogen atom or a C1-C8 alkyl group,  $R_3$  represents a C1-C4 alkylidene group, A represents a metal having a valence of (n+x), n is an integer of 1 to 3, and x is an integer of 0 to 2).

[0007]

The present invention will next be described in detail. [0008]

(Phosphoric acid aromatic ester metal salt)

Examples of the C4-C8 alkyl group represented by  $R_1$  of formula (I) include butyl, sec-butyl, tert-butyl, pentyl, tert-pentyl, hexyl, cyclohexyl, heptyl, octyl, isooctyl, and tert-octyl.

[0009]

Examples of the alkyl group represented by  $R_2$  include methyl, ethyl, propyl, isopropyl, butyl, sec-butyl, tert-butyl, pentyl, tert-pentyl, hexyl, cyclohexyl, heptyl, octyl, isooctyl, and tert-octyl.

[0010]

Examples of the alkylidene group represented by  $R_3$  include methylene, ethylidene, propylidene, and butylidene. [0011]

Examples of the metal represented by A include alkali metals such as lithium, sodium, and potassium; alkaline earth metals such as magnesium and calcium; zinc and aluminum.

[0012]

The phosphoric acid aromatic ester metal salt of the present invention having a certain average particle size, certain average aspect ratio, and certain bulk specific gravity which individually fall within specified ranges can be produced by pulverizing a phosphoric acid aromatic ester metal salt synthesized through a conventionally known method by use of a pulverization apparatus such as a ball mill, a Henschel mixer, or a kneader.

[0013]

Specific examples of the compound represented by formula (I) include the following compounds No.1 to 4. However, the present invention is not limited to the following compounds.

[0014]

[Chemical Formula 3]

Compound No.1

$$\begin{array}{c|c} t\text{-}C_4H_9 \\ \hline \\ t\text{-}C_4H_9 \\ \hline \\ t\text{-}C_4H_9 \\ \hline \\ t\text{-}C_4H_9 \\ \hline \end{array}$$

[0015]

[Chemical Formula 4]

Compound No.2

$$CH_3$$
 $CH_2$ 
 $P-O$ 
 $Na^+$ 
 $CH_3$ 
 $CH_3$ 
 $CH_4$ 
 $CH_5$ 
 $CH_5$ 
 $CH_6$ 
 $C$ 

[0016]

[Chemical Formula 5]

Compound No.3

$$t-C_5H_{11}$$
 $CH_3-CH$ 
 $P-O$ 
 $Na^+$ 
 $t-C_5H_{11}$ 

[0017]

[Chemical Formula 6]

Compound No.4

$$\begin{array}{c|c} & \text{t-C}_5H_{11} \\ & & \\ &$$

[0018]

[Chemical Formula 7]

Compound No.5

$$\begin{bmatrix}
t-C_4H_9 & & & \\
t-C_4H_9 & & & & \\
CH_2 & P-O & Ca \\
t-C_4H_9 & & & & \\
t-C_4H_9 & & & & \\
\end{bmatrix}$$

[0019]

[Chemical Formula 8]

Compound No.6

$$\begin{bmatrix} t-C_4H_9 & & & \\ t-C_4H_9 & & & & \\ & & CH_2 & P-O & Al-OH \\ t-C_4H_9 & & & & \\ & & t-C_4H_9 & & & \\ \end{bmatrix}$$

[0020]

As described above, no particular limitation is imposed on the method for synthesizing the phosphoric acid aromatic ester metal salt represented by the aforementioned formula

(I), and the metal salt is synthesized through a known method. The nucleating agent of the present invention having a certain average particle size, a certain average aspect ratio, and a certain bulk specific gravity which individually fall within specified ranges is produced by pulverizing the metal salt for a predetermined period of time, by means of a combination of pulverization methods or a specific pulverization method.

[0021]

Specific examples of the pulverization apparatus used for producing the nucleating agent of the present invention include compression pulverization apparatuses such as a brake crusher, a dodge crusher, a single-toggle-jaw crusher, a gyratory crusher, a cone crusher, a hydro-cone crusher, a roll crusher, a single-roll crusher, and a disk crusher; impact-compression pulverization apparatuses such as a stamp mill, a hammer mill, an impeller breaker, an impact crusher, a Raymond vertical mill, a disintegrator, a dismembrater, a titanium mill, a novorotor, a micron mill, a jet mill, a jet pulverizer, a micronizer, a reductionizer, a jet pulverization apparatus, an air mill, a ball mill, a tube mill, a rod mill, a conical mill, a tricone mill, and a Hildebrand mill; shear pulverization apparatuses such as a cutting mill, a rotary crusher, and a shear roll mill; and friction pulverization apparatuses such as a hand mill, a pan mill, an attrition mill, an edge runner, a sand grinder, a screw crusher, a pillar-shaped attrition mill, a centrifugal

roller mill, a centrifugal ball mill, a ring roller mill, a high-speed ball mill, a low-speed ball mill, a high-swing ball mill, a premier mill, and a Schalotte mill.

Particularly, combined use of two or more different types of pulverization apparatuses is preferred.

[0022]

As described below in Examples and Comparative Examples, powder of particles having a small average particle size and a large aspect ratio or powder of particles having a small average particle size and a small aspect ratio is obtained by appropriately selecting a specific pulverization method and a certain pulverization time, or by means of a combination of pulverization methods and times.

[0023]

Examples of crystalline synthetic resins into which the nucleating agent of the present invention is to be incorporated include  $\alpha$ -olefin polymers such as polypropylene, high density polyethylene, low density polyethylene, linear-chain low density polyethylene, polybutene-1, poly(3-methylpentene), poly(4-methylpentene), and ethylene/propylene block or random copolymers; thermoplastic linear-chain polyesters such as polyethylene terephthalate, polybutylene

The amount of the nucleating agent of the present invention to be incorporated into a crystalline synthetic

polyphenylene sulfide; polycaprolactone; and linear-chain

terephthalate, and polyhexamethylene terephthalate;

polyamides such as polyhexamethylene adipamide.

resin varies in accordance with the type of the resin. The amount of incorporation is typically 0.001-10 parts by weight, preferably 0.01-5 parts by weight, on the basis of 100 parts by weight of the resin.

[0024]

When the nucleating agent of the present invention is incorporated into, for example,  $\alpha$ -olefin polymer of the aforementioned resins, the effects of the nucleating agent vary in accordance with the density, softening point, melt flow rate, and molecular weight distribution of the polymer; the percentage of components of the polymer which are insoluble in a solvent; the degree of stereospecificity of the polymer; the type of a polymerization catalyst; whether or not the catalyst residue is treated; and the type of olefins serving as raw materials and the compositional proportions thereof. However, the nucleating agent of the present invention is much or less effective for any of the aforementioned resins. Examples of the resin into which the nucleating agent is to be incorporated include resins and resin compositions described in Japanese Patent Application Laid-Open (kokai) No. S63-37148, S63-37152, S63-90552, S63-210152, S63-213547, S63-243150, S63-243152, S63-260943, S63-260944, S63-264650, H01-178541, H02-49047, H02-102242, H02-251548, Н02-279746, Н03-195751, Н07-118466, Н07-118486, Н07-133380, H07-138421, H07-173302, H07-173317, H07-173341, H07-179684, Н07-179691, Н07-195592, Н07-238204, Н07-242776, Н07-268143, Н07-286089, Н08-27335, Н08-34885, Н08-59920, Н0859921, H08-73671, H08-81589, H08-81595, H08-134293, H08-269266, H08-291236, H08-311272, H08-311295, H08-231788, H09-3274, H09-3293, H09-12805, H09-20840, H09-59443, H09-59455, and H09-67501.

[0025]

The nucleating agent of the present invention may contain a mixture of two or more different phosphoric acid aromatic ester metal salts, so long as the average particle size, etc. of the mixture falls within the range specified by the present invention. If necessary, the nucleating agent is preferably used in combination with, for example, an antioxidant such as a phenolic antioxidant, a phosphorus-containing antioxidant, or a thioether-containing antioxidant, a UV absorber, a hindered amine photo-stabilizer, another nucleating agent, an antistatic agent, a filler, a flame retardant, or a lubricant.

[0026]

Examples of the aforementioned phenolic antioxidant include 2,6-di-tert-butyl-p-cresol, 2,6-diphenyl-4-octadecyloxyphenol, distearyl (3,5-di-tert-butyl-4-hydroxybenzyl)phosphonate, 1,6-hexamethylenebis[(3,5-di-tert-butyl-4-hydroxyphenyl)propionic acid amide], 4,4'-thiobis(6-tert-butyl-m-cresol), 2,2'-methylenebis(4-methyl-6-tert-butylphenol), 2,2'-methylenebis(4-ethyl-6-tert-butylphenol), 4,4'-butylidenebis(6-tert-butyl-m-cresol), 2,2'-ethylidenebis(4-fert-butylphenol), 2,2'-ethylidenebis(4-fert-butylphenol), 1,1,3-tris(2-methyl-4-hydroxy-fert-butylphenol), 1,1,3-tris(2-methyl-4-hydroxy-fert-butylphenol), 1,1,3-tris(2-methyl-4-hydroxy-fert-butylphenol), 1,1,3-tris(2-methyl-4-hydroxy-fert-butylphenol), 1,1,3-tris(2-methyl-4-hydroxy-fert-butylphenol), 1,1,3-tris(2-methyl-4-hydroxy-fert-butylphenol), 1,1,3-tris(2-methyl-4-hydroxy-fert-butylphenol), 1,1,3-tris(2-methyl-4-hydroxy-fert-butylphenol)

5-tert-butylphenyl)butane, 1,3,5-tris(2,6-dimethyl-3-hydroxy-4-tert-butylbenzyl) isocyanurate, 1,3,5-tris(3,5-di-tertbutyl-4-hydroxybenzyl) isocyanurate, 1,3,5-tris(3,5-di-tertbutyl-4-hydroxybenzyl)-2,4,6-trimethylbenzene, 2-tert-butyl-4-methyl-6-(2-acryloyloxy-3-tert-butyl-5-methylbenzyl)phenol, stearyl (3,5-di-tert-butyl-4-hydroxyphenyl) propionate, thiodiethylene glycol bis[(3,5-di-tert-butyl-4hydroxyphenyl)propionate], 1,6-hexamethylenebis[(3,5-di-tertbutyl-4-hydroxyphenyl)propionate], bis[3,3-bis(4-hydroxy-3tert-butylphenyl)butyric acid] glycol ester, bis[2-tertbutyl-4-methyl-6-(2-hydroxy-3-tert-butyl-5methylbenzyl)phenyl] terephthalate, 1,3,5-tris[(3,5-di-tertbutyl-4-hydroxyphenyl)propionyloxyethyl] isocyanurate, tetrakis[methyl 3-(3,5-di-tert-butyl-4hydroxyphenyl)propionate]methane, 3,9-bis[1,1-dimethyl-2-{(3tert-butyl-4-hydroxy-5-methylphenyl)propionyloxy}ethyl]-2,4,8,10-tetraoxaspiro[5.5]undecane, and triethylene glycol bis[(3-tert-butyl-4-hydroxy-5-methylphenyl)propionate]. phenolic antioxidant is incorporated into a resin in an amount of 0.001-10 parts by weight, preferably 0.05-5 parts by weight, on the basis of 100 parts by weight of the resin. [0027]

Examples of the aforementioned phosphorus-containing antioxidant include tris(nonylphenyl) phosphite, tris(2,4-di-tert-butylphenyl) phosphite, tris[2-tert-butyl-4-(3-tert-butyl-4-hydroxy-5-methylphenylthio)-5-methylphenyl] phosphite, tridecyl phosphite, octyldiphenyl phosphite,

di(decyl)monophenyl phosphite, di(tridecyl)pentaerythritol diphosphite, di(nonylphenyl)pentaerythritol diphosphite, bis(2,4-di-tert-butylphenyl)pentaerythritol diphosphite, bis(2,6-di-tert-butyl-4-methylphenyl)pentaerythritol diphosphite, bis(2,4,6-tri-tert-butylphenyl)pentaerythritol diphosphite, bis(2,4-dicumylphenyl)pentaerythritol diphosphite, tetra(tridecyl)isopropylidenediphenol diphosphite, tetra(tridecyl)-4,4'-n-butylidenebis(2-tertbutyl-5-methylphenol) diphosphite, hexa(tridecyl)-1,1,3tris(2-methyl-4-hydroxy-5-tert-butylphenyl)butane triphosphite, tetrakis(2,4-di-tert-butylphenyl)biphenylene diphosphonite, 9,10-dihydro-9-oxa-10-phosphaphenanthrene-10oxide, 2,2'-methylenebis(4,6-tert-butylphenyl)-2-ethylhexyl phosphite, 2,2'-methylenebis(4,6-tert-butylphenyl)octadecyl phosphite, 2,2'-ethylidenebis(4,6-di-tert-butylphenyl) fluorophosphite, tris(2-[(2,4,8,10-tetrakis(tertbutyl)dibenzo[d,f][1,3,2]dioxaphosphepin-6-yl)oxy]ethyl)amine, and a phosphite of 2-ethyl-2-butylpropylene glycol and 2,4,6tri-tert-butyl phenol.

[0028]

Examples of the thioether-containing antioxidant include dialkyl thiodipropionates such as dilauryl thiodipropionate, dimyristyl thiodipropionate, and distearyl thiodipropionate; and  $\beta$ -alkylmercaptopropionic acid esters of polyol such as pentaerythritol tetra( $\beta$ -dodecylmercaptopropionate).

[0029]

Examples of the UV absorber include 2hydroxybenzophenones such as 2,4-dihydroxybenzophenone, 2hydroxy-4-methoxybenzophenone, 2-hydroxy-4-octoxybenzophenone, and 5,5'-methylenebis(2-hydroxy-4-methoxybenzophenone); 2-(2'-hydroxyphenyl)benzotriazoles such as 2-(2'-hydroxy-5'methylphenyl)benzotriazole, 2-(2'-hydroxy-3',5'-di-tertbutylphenyl)-5-chlorobenzotriazole, 2-(2'-hydroxy-3'-tertbutyl-5'-methylphenyl)-5-chlorobenzotriazole, 2-(2'-hydroxy-5'-tert-octylphenyl)benzotriazole, 2-(2'-hydroxy-3',5'dicumylphenyl)benzotriazole, 2,2'-methylenebis(4-tert-octyl-6-benzotriazolyl)phenol, and 2-(2'-hydroxy-3'-tert-butyl-5'carboxyphenyl)benzotriazole; benzoates such as phenyl salicylate, resorcinol monobenzoate, 2,4-di-tert-butylphenyl 3,5-di-tert-butyl-4-hydroxybenzoate, 2,4-di-tert-amylphenyl 3,5-di-tert-butyl-4-hydroxybenzoate, and hexadecyl 3,5-ditert-butyl-4-hydroxybenzoate; substituted oxanilides such as 2-ethyl-2'-ethoxyoxanilide and 2-ethoxy-4'-dodecyloxanilide; cyanoacrylates such as ethyl  $\alpha$ -cyano- $\beta$ ,  $\beta$ -diphenylacrylate and methyl 2-cyano-3-methyl-3-(p-methoxyphenyl)acrylate; and triaryltriazines such as 2-(2-hydroxy-4-octoxyphenyl)-4,6bis(2,4-di-tert-butylphenyl)-s-triazine, 2-(2-hydroxy-4methoxyphenyl)-4,6-diphenyl-s-triazine, and 2-(2-hydroxy-4propoxy-5-methylphenyl)-4,6-bis(2,4-di-tert-butylphenyl)-striazine.

[0030]

Examples of the hindered amine photo-stabilizer include hindered amine compounds such as 2,2,6,6-tetramethyl-4-

piperidyl stearate, 1,2,2,6,6-pentamethyl-4-piperidyl stearate, 2,2,6,6-tetramethyl-4-piperidyl benzoate, bis (2,2,6,6-tetramethyl-4-piperidyl) sebacate, bis (1,2,2,6,6-tetramethyl-4-piperidyl)pentamethyl-4-piperidyl) sebacate, tetrakis(2,2,6,6tetramethyl-4-piperidyl) butanetetracarboxylate, tetrakis(1,2,2,6,6-pentamethyl-4-piperidyl) butanetetracarboxylate, bis(2,2,6,6-tetramethyl-4piperidyl) ·di(tridecyl) -1,2,3,4-butanetetracarboxylate, bis  $(1,2,2,6,6-pentamethyl-4-piperidyl) \cdot di(tridecyl)-1,2,3,4$ butanetetracarboxylate, bis(1,2,2,6,6-pentamethyl-4piperidyl)-2-butyl-2-(3,5-di-tert-butyl-4-hydroxybenzyl) malonate, 1-(2-hydroxyethyl)-2,2,6,6-tetramethyl-4piperidinol/diethyl succinate polycondensates, 1,6bis(2,2,6,6-tetramethyl-4-piperidylamino)hexane/dibromoethane polycondensates, 1,6-bis(2,2,6,6-tetramethyl-4piperidylamino) hexane/2, 4-dichloro-6-morpholino-s-triazine polycondensates, 1,6-bis(2,2,6,6-tetramethyl-4piperidylamino) hexane/2, 4-dichloro-6-tert-octylamino-striazine polycondensates, 1,5,8,12-tetrakis[2,4-bis(N-butyl-N-(2,2,6,6-tetramethyl-4-piperidyl)amino)-s-triazin-6-yl]-1,5,8,12-tetraazadodecane, 1,5,8,12-tetrakis[2,4-bis(N-butyl-N-(1,2,2,6,6-pentamethyl-4-piperidyl)amino)-s-triazin-6-yl]-1,5,8,12-tetraazadodecane, 1,6,11-tris[2,4-bis(N-butyl-N-(2,2,6,6-tetramethyl-4-piperidyl)amino)-s-triazin-6yl]aminoundecane, and 1,6,11-tris[2,4-bis(N-butyl-N-(1,2,2,6,6-pentamethyl-4-piperidyl)amino)-s-triazin-6yl]aminoundecane.

[0031]

Examples of other nucleating agents include metal salts of benzoic acid such as aluminum p-tert-butylbenzoate and lithium p-tert-butylbenzoate; benzylidene sorbitols such as dibenzylidene sorbitol, bis(4-methylbenzylidene) sorbitol, bis(4-ethylbenzylidene) sorbitol, and bis(dimethylbenzylidene) sorbitol; metal alcoholates such as zinc glyceride; and amino acid metal salts such as zinc glutamate.

[0032]

A synthetic resin composition containing the nucleating agent of the present invention can be used in a variety of fields including construction materials, agricultural materials, automobile parts, packaging materials, sundries, toys, and electric appliances, in accordance with properties of the synthetic resin contained in the composition.

[0033]

[Examples]

(Synthesis of Compound No. 1)

2,2'-Methylenebis(4,6-di-t-butylphenyl) phosphate (1,458 g, 3 mol), sodium hydroxide (120 g, 3 mol) in water (300 g), and methanol (292 g) were added to a kneader, and the resultant mixture was kneaded at room temperature for one hour. The resultant product was dried under vacuum, and then pulverized, to thereby yield white powdery compound No. 1 (1,264 g). The thus-yielded powdery compound No. 1 had an average particle size of 27  $\mu$ m, an aspect ratio of 16, and a

bulk specific gravity of 0.08. The powder as a raw material powder A was subjected to the below-described pulverization test.

[0034]

(Pulverization test)

Raw material powder A was pulverized by use of a pulverization apparatus shown in Table-1 for 30 minutes or three hours, and the average particle size, aspect ratio, and bulk specific gravity of the pulverized product were measured. Furthermore, in order to confirm the effect of combination of pulverization methods, raw material powder A was pulverized by means of two different pulverization methods (for 30 minutes each); or raw material powder A was pulverized by means of one of the two methods for 30 minutes and then by means of the other method for two hours and 30 minutes. respective cases, the average particle size, aspect ratio, and bulk specific gravity of the pulverized product were measured. The results are shown in Table 1. The pulverization methods are as follows: method 1: pulverization by use of a jet mill, method 2: pulverization by use of a ball mill.

The average particle size and the bulk specific gravity (bulk density) were measured in accordance with JIS K6900. The average aspect ratio was obtained as follows: the lengths of the major axes and minor axes of randomly chosen 100 crystals were measured from a micrograph of the above-pulverized powder, and the average of the ratio of the major

axis lengths to the minor axis lengths was obtained. [0035]

[Table 1]

Table 1

|                | Pulverization method |      |        | Average<br>particle<br>size | Average<br>Aspect | Bulk<br>specific<br>gravity |       |
|----------------|----------------------|------|--------|-----------------------------|-------------------|-----------------------------|-------|
|                | Method               | Time | Method | Time                        | μm                | ratio                       | g/cm³ |
| Comp.<br>Ex. 1 | 1                    | 0.5  | _      |                             | 3.2               | 26                          | 0.06  |
| Comp.<br>Ex. 2 | 1                    | 3.0  | _      |                             | 1.8               | 23                          | 0.08  |
| Comp.<br>Ex. 3 | 2                    | 0.5  | _      |                             | 2.5               | 15                          | 0.12  |
| Ex. 1          | 2                    | 3.0  | _      |                             | 1.7               | 7                           | 0.15  |
| Ex. 2          | 1                    | 0.5  | 2      | 0.5                         | 1.8               | 9                           | 0.13  |
| Ex. 3          | 1                    | 0.5  | 2      | 2.5                         | 0.3               | 2                           | 0.25  |
| Ex. 4          | 2                    | 0.5  | 1      | 2.5                         | 0.6               | 6                           | 0.19  |

[0036]

The results of Comparative Examples 1 and 2, Example 1 and Comparative Example 3 show that, when pulverization time is lengthened, the average particle size is reduced regardless of the pulverization method, and that the aspect ratio is considerably reduced or slightly reduced in accordance with the pulverization method employed, even when pulverization time is lengthened. Accordingly, when pulverization is carried out by means of an appropriately selected method for a long period of time, powder having a small aspect ratio can be obtained. The results of Examples 2 and 3 show that, when pulverization methods are carried out in combination, powder having a small aspect ratio can be obtained effectively.

[0037]

## (Reference Example)

Effects of the nucleating agent of the present invention when incorporated into a resin will next be described by way of Reference Examples, which should not be construed as limiting the invention thereto.

[0038]

Polypropylene ("Profax 6501", Montell) (100 parts by weight), calcium stearate (0.05 parts by weight), tetrakis[methyl 3-(3,5-di-tert-butyl-4-hydroxyphenyl)propionate]methane (0.1 parts by weight), and a nucleating agent (shown in Table 2) (0.1 parts by weight) were blended by use of a Henschel mixer, and the resultant mixture was extruded at 250°C, to thereby form pellets. The resultant pellets were subjected to injection molding at 230°C, to thereby produce sheet-like test pieces. The haze and Izod impact resistance at 20°C (kg·cm/cm²) of the thusproduced test piece were measured in accordance with ASTM D-1003-61 and ASTM D-256, respectively. The results are shown in Table 2.

[0039]

[Table 2]

Table 2

| Reference Example                | Nucleating agent | Haze | Izod impact resistance |
|----------------------------------|------------------|------|------------------------|
| 1-1                              | Example-1        | 15   | 6.6                    |
| 1-2                              | Example-2        | 14   | 6.5                    |
| 1-3                              | Example-3        | 12   | 6.8                    |
| 1-4                              | Example-4        | 13   | 6.8                    |
| Comparative<br>Reference Example |                  |      |                        |
| 1-1                              | Comp. Example-1  | 20   | 6.1                    |
| 1-2                              | Comp. Example-2  | 21   | 6.2                    |
| 1-3                              | Comp. Example-3  | 23   | 6.0                    |

[0040]

[Effect of the Invention]

When a phosphoric acid aromatic ester metal salt nucleating agent is pulverized so as to attain an average particle size of 10 µm or less, an aspect ratio of 10 or less, and a bulk specific gravity of at least 0.1, the resultant nucleating agent exhibits excellent handling property (i.e., chargeability), suppresses generation of dust, and exerts excellent effects of improving properties of a crystalline synthetic resin when incorporated into the resin.

[Name of Document] Abstract
[Abstract]

[Problems to be solved] The nucleating agent exhibits excellent handling property when incorporated into a resin, and is highly effective in improving transparency and mechanical strength of the resin containing the agent.

[Means to solve the Problem] A nucleating agent containing a phosphoric acid aromatic ester metal salt represented by formula (I) and having an average particle size of 10 µm or less, an average aspect ratio of 10 or less, and a bulk specific gravity of 0.1 or more:

[Chemical Formula 1]

$$\begin{bmatrix} R_1 & & & & \\ R_2 & & & & \\ R_3 & & P-O \\ R_2 & & & \\ R_1 & & & \\ \end{bmatrix}_n A(OH)x \qquad (I)$$

(wherein  $R_1$  represents a C4-C8 alkyl group,  $R_2$  represents a hydrogen atom or a C1-C8 alkyl group,  $R_3$  represents a C1-C4 alkylidene group, A represents a metal having a valence of (n+x), n is an integer of 1 to 3, and x is an integer of 0 to 2).

[Chosen Drawing] None